

Electrohydrodynamic instability in a mixture of cyanobiphenyl and cyanoterphenyl

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Abstract : Electrohydrodynamic (EHD) instability in a homeotropically oriented liquid crystal mixture of cyanobiphenyl and terphenyl, having positive dielectric anisotropy, has been studied under a dc electric field. The sample has no sharp phase transition temperature. Smectic and nematic phases, nematic and isotropic phases were seen to co-exist. Effects of electric field on two different phases were observed simultaneously. Studies were carried out on liquid crystal cells of four different thicknesses. Textures and electrical properties were studied at different voltages and temperatures. Existence of charge injection, charge diffusion and convective flow of liquid in the liquid crystal cells were found. Role of viscosity, dielectric anisotropy and other physical parameters on electric current through the sample cells, were discussed.

Keywords : Electrohydrodynamic instability, liquid crystal mixture, charge diffusion

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1. Introduction

Onset of field-induced hydrodynamic instability, Electrohydrodynamic (EHD) instability, has been studied by many workers. In nematic liquid crystals with negative dielectric anisotropy EHD instability was observed by Williams and his co-workers [1–3]. In nematic liquid crystals with positive dielectric anisotropy EHD was first observed in 5CB by Nakagawa [4]. Rout and Choudhary [5,6] extensively studied EHD instability in 8CB and 8OCB. We have studied EHD instability in binary mixtures of cyanobiphenyls [7,8] in a dc electric field. This instability in a dc electric field was explained by the Felici-Benard instability [FBI] model [5,6]. In this model charges are injected into liquid crystal from the electrodes. Interaction between the injected charges and applied electric field induces a

vortical flow in the liquid crystal confined between the electrodes causing the appearance of roll-like domain patterns under the polarizing microscope. It also results in cellular patterns in the charge and velocity distribution in the plane perpendicular to the applied electric field [9]. After the occurrence of EHD instability, the current through the liquid crystal consists of three main terms [9], the ohmic term, diffusion term, and convection term arises from the convective flow of liquid in the liquid crystal accompanied by charge transfer. Liquid crystals possess anisotropies in different physical parameters which should have a great influence on the fluid flow, consequently EHD instability in a nematic liquid crystal could be influenced by the said anisotropies. In our present study, we have taken a mixture sample S_3 , kindly supplied by M/S BDH Ltd (UK). The EHD instability in S_3 , has not been reported yet. As reported by BDH, this mixture has no sharp phase transition temperature. For this reason, the coexistence of smectic and nematic phases and nematic and isotropic phases were found in the mixture. The electric field effects on a liquid crystal in different phases can be observed simultaneously.

2. Experimental

For the present study we received a mixture of cyanobiphenyl and terphenyl, called as S_3 , as a gift from M/S BDH Ltd (UK). For electrical studies, liquid crystal cells were prepared by sandwiching the liquid crystal mixture between two Sb doped SnO_2 coated glass plates working as electrodes. The effective electrode area was pre-determined by etching the conducting layer (8 mm \times 8 mm). To ensure the uniformity of sample thickness, optically plane glass plates and standard Mylar spacers of known thickness were used. Liquid crystal cells with four different thicknesses, 17 μm , 23 μm , 50 μm and 75 μm were prepared. Mettler central processor (FP-800) and a hot stage (FP-82) were used to control and measure the temperature of the sample with a temperature resolution of 0.1°C. By pretreating the glass plates with CTAB (cetyl trimethyl ammonium bromide), homeotropic alignment of the liquid crystal molecules was obtained. The visual observations and microphotographic work were done by the polarizing microscope. A set of dry cells and a potentiometer were used for supply and control of the dc voltage applied across the cells. The current through the cells was measured by a pico-ammeter (DPM-111). Studies on charge diffusion through the sample were done by switching on and off a dc voltage across the cell and recording the transient current, after switching off the applied voltage, by a digital memoryscope (Iwatsu DMS-6430) with a data length of 1024 wd.

3. Results and discussion

A. Microscopic studies :

Microscopic studies were carried out with keeping the polarizer and analyzer of microscope in crossed position. From the study of textures we obtained the following sequence of phase transitions,

$$\text{SmA} \leftarrow 53.3^\circ\text{C} - 53.8^\circ\text{C} \rightarrow \text{N} \leftarrow 55.6^\circ\text{C} - 57.8^\circ\text{C} \rightarrow \text{I},$$

where SmA = smectic phase, N = nematic phase, I = isotropic phase.

(i) Smectic A phase :

In zero field condition, in the smectic A phase, for all the four cells, focal conic textures were observed. At any temperature below 53°C, a dc voltage, up to 10 V, applied to the sample did not show any change in the liquid crystal textures. At temperature 53.2°C, with applied voltage 3.5 V, a change in the texture was observed. In some micro regions, focal conic textures disappeared and a dark texture appeared. The area of the dark micro-regions was found to increase with the increasing of the applied voltage. In thin cells, this type of occurrence was observed very clearly but in the thick cells, most of the area under the field of view remained same with focal conic textures. On removal of the electric field, area of the dark regions was found reduced but did not returned to its initial focal conic textures completely. The threshold voltage, V_{thf} , of this type of transition was observed very much temperature sensitive near the SmA-N transition temperature.

(ii) Smectic A and Nematic phase :

At 53.3°C, phase transition SmA-N started. In micro-regions, where focal conic to dark texture transition was observed previously on the application of the electric field, the phase transition started first. For the temperature range, 53.3°C–53.8°C, the liquid crystal was found to exist both in the nematic phase and smectic phase interspersed through out the cell. The nematic area was found to increase with the increase of the temperature. The texture of the nematic area appeared dark due to homeotropic alignment of the molecules. A few bright spots were still found with in the nematic zone, specially in the thick cells showing a lack of proper alignment at those spots. When a voltage of 2 V was applied, the bright spots in nematic area became dark due to completion of homeotropic alignment. Initially, in the temperature range 53.3°C–53.8°C, focal conic textures were found in the smectic A zone but the textures were not so sharp in appearance. Above an applied voltage of 2.5 V, the transition from focal conic to dark texture was found in the smectic A zone also. Initially this transition started at the smectic-Nematic boundaries and gradually spread through out the total smectic area with the increase of the applied voltage but a considerable number of bright spots still existed which were the residues of the focal conic texture. At this time, if the electric field was withdrawn, the size of the spots increased and the focal conic textures were again visible in those micro-regions but the rest of the smectic zone remained dark and the alignment of the molecules in those micro-regions was retained homeotropic.

In the liquid crystals with positive dielectric anisotropy ($\Delta\epsilon > 0$), Freedericksz transition for planer to homeotropic alignment is possible if an electric field is applied along the cell thickness. The threshold voltage depends on the splay elastic constant and dielectric anisotropy [$V_{th} = C(K_{11}/\Delta\epsilon)^{1/2}$, where V_{th} = threshold voltage for above mentioned Freedericksz transition, C = constant, K_{11} = splay elastic constant] [10]. From the above observations it is clear that the liquid crystal mixture consists of positive dielectric anisotropy and the splay elastic constant of the liquid crystal in smectic phase is higher than in the nematic phase.

When the applied voltage was further increased gradually the cellular domain patterns started to appear in the nematic zone. At temperature 53.6°C , the threshold voltage V_{th} was found to be 4 V for the $17\text{ }\mu\text{m}$ cell and 3.8 V for the $50\text{ }\mu\text{m}$ cell. Figures 1(a)–1(c) are the microphotographs for the $17\text{ }\mu\text{m}$ cell at 4.5 V, 5.5 V and 6 V respectively. In Figure 1(a), EHD instability started in one corner which was in nematic phase but the area in the other corner (which the liquid crystal was still in smectic phase) containing bright spots remained unchanged. With increasing applied voltage, the bright spots disappeared and gradually EHD instability was observed to appear through out the total cell area [Figures 1(b), 1(c)]. The liquid crystal under the smectic phase behaved like a nematic liquid crystal and domain patterns appeared. But, for the $75\text{ }\mu\text{m}$ cell at 53.4°C , EHD instability was not seen to spread over the smectic area with the increasing applied voltage [Figure 1(k)].

Figures 1(d–f) are microphotographs of $50\text{ }\mu\text{m}$ cell at 53.6°C and in different voltages. The cellular domains appeared at 3.8 V and elongated along with increasing voltage. Figure 1(d) shows the domain pattern at 4.2 V, where a few irregular bright spots still exist. At this time the flow of odd impurities present in the sample was observed clearly in the roll-like domains and the flow direction were opposite to each other in the neighboring domains which make it clear that the appearance of domains is a indication of convective liquid flow. At 4.6 V the length of the roll-like domains began to contract but increase in number [Figure 1(e) for 5 V]. At 6 V, the liquid flow increased and rolls converted into a large number of droplet like domains [Figure 1(f)]. Above 6.5 V the domains started to distort and the liquid flow became turbulent above 7 V. With further increase of the applied voltage, the regularity in fluid flow was destroyed completely.

(iii) Nematic phase :

In the temperature range 53.8°C – 55.6°C , the sample was in the nematic phase. Figures 1(g) and 1(h) are the photographs of $75\text{ }\mu\text{m}$ cell at 5 V and 6 V respectively at temperature 54°C . Here also the cell appeared dark initially due to the homeotropic alignment of the liquid crystal molecules and with increasing applied voltage well organized roll-like domains appeared. Up to 5 V, the rolls were found to elongate and above it the roll lengths contracted gradually but the number of separate domains increased. It was due to the fact that, at this time, the rate of fluid flow increased and the long cylindrical vortex broke into separate closed tube-like vertical flow giving rise of above observations. Here also, the fluid flow became turbulent at 7 V.

(iv) Nematic and isotropic phase :

The nematic phase and isotropic phase were seen to coexist in the temperature range 55.6°C – 57.8°C . Figures 1(i) and 1(j) show the microphotographs taken at 4V and at temperatures 56.5°C and 56.9°C respectively. Here the nematic phase (area covered with roll-like domains) and the isotropic phase (dark circular area) coexist. At 56.5°C , V_{th} was found to be 3.4 V and at 56.9°C it was 3.5 V. The cellular domains appeared similarly as discussed above. The domain patterns started appearing at the area far from the isotropic zone. This was due to the fact that for the nematic liquid crystal in the vicinity of the



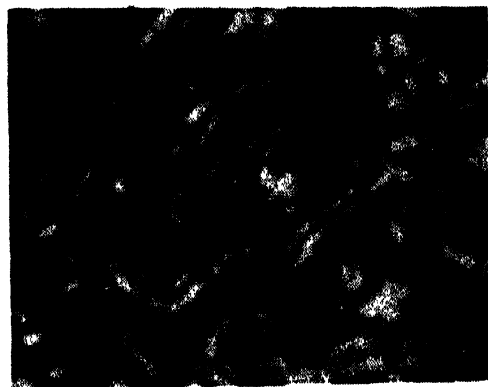
1(a)



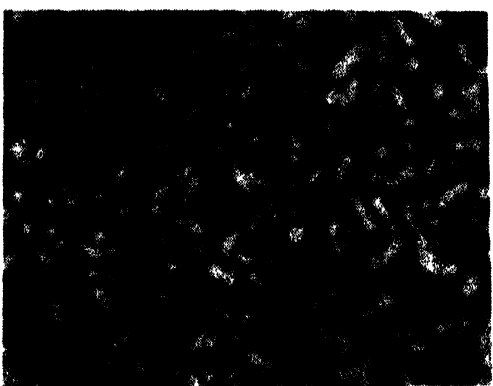
1(b)



1(c)



1(d)



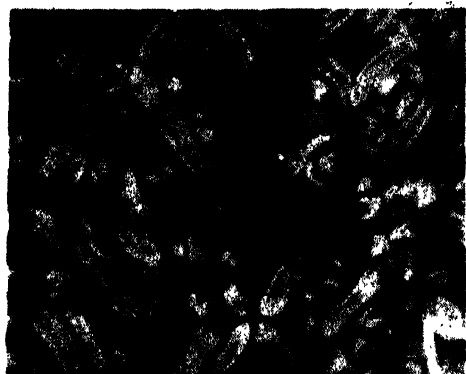
1(e)



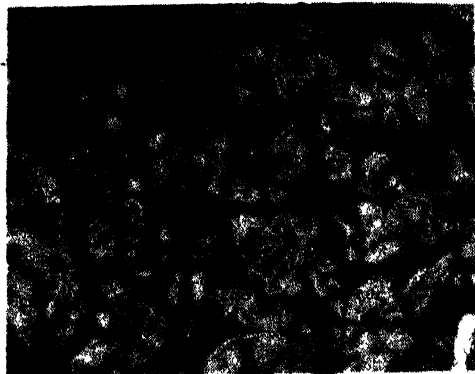
1(f)

Figure 1. Microphotographs of domain patterns in sample 'S₃' (magnification 150 ×). For the 17 μm thick cell with applied voltage 1(a) 4.8 V, 1(b) 5.5 V, 1(c) 6 V. For the 50 μm thick cell with applied voltage 1(d) 4.12 V, 1(e) 5 V, 1(f) 6 V, at 53.6°C.

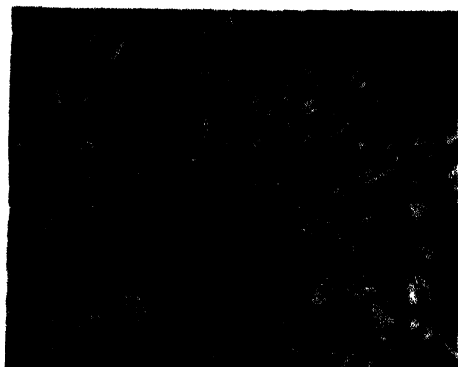
Plate I (Cont'd.)



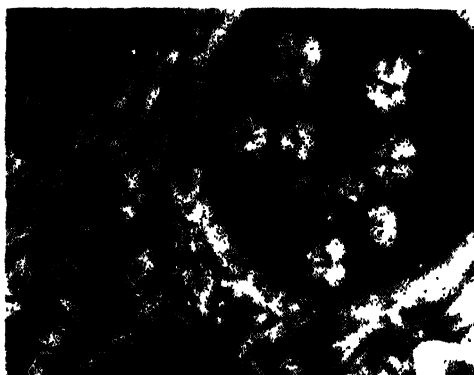
1(g)



1(h)



1(i)



1(j)



1(k)

Figure 1. Microphotographs of domain patterns in sample 'S₃' (magnification 150 ×). For the 75 μm thick cell with applied voltage 1(g) 5 V, 1(h) 6 V at 54°C and 1(i) 4 V at 56.5°C, 1(j) 4.2 V at 56.9°C, 1(k) 4.2 V at 53.4°C.

isotropic zone, $(T_{N-I} - T)$ was very close to zero [T = working temperature, T_{N-I} = nematic to isotropic phase transition temperature], so the order parameter was very small [11]. The thermal vibrations of the directors about their mean direction increased highly. To make the optical patterns visible, a voltage was required to be applied which was higher than the threshold voltage, V_{th} [12] necessary to start EHD instability in the liquid crystal at that respective area. But in the areas remote from the isotropic zone, the optical patterns appeared with the starting of EHD instability almost simultaneously. Another interesting observation was that, within the isotropic liquid nematic droplets were seen to appear on starting of EHD instability. Due to vortical flow in the liquid crystal, small amount of nematic liquid crystal might be transfused into the isotropic liquid and formed the droplets. In figure 1(j), a few faint crosses appeared in the isotropic zone.

B. Electrical measurements :

(i) Current-voltage characteristics :

Figures 2(a) and (b) show the variation of current (I) with voltage (V) at different temperatures (T) for the four cells of different thicknesses. When the voltage was below 2 V,

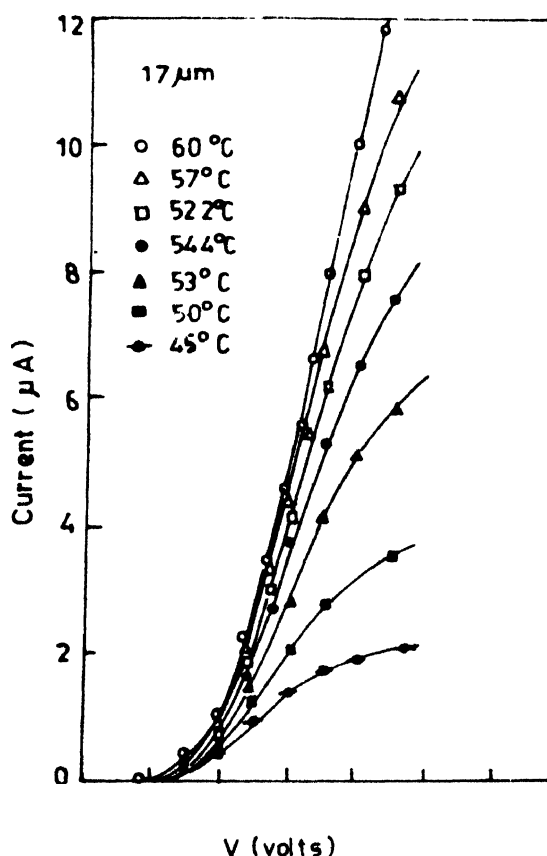


Figure 2(a). Current-voltage characteristics at different temperatures for 'S₃' for cell thickness 17 μ m.

a very small current was found to flow. It was due to ohmic conduction of current through the liquid crystal. Above 2 V, both in smectic phase and in nematic phase, the current

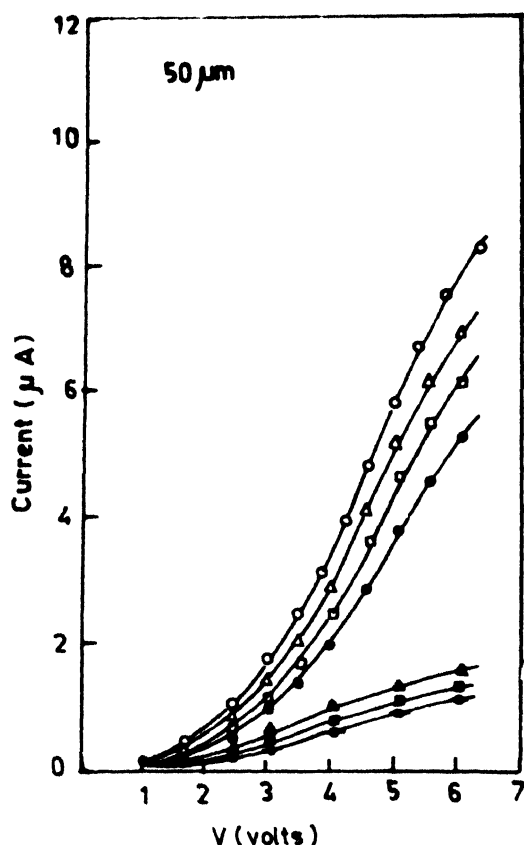


Figure 2(b). Current-voltage characteristics at different temperatures for 'S₃' for cell thickness 50 μm.

increases with a higher rate with increasing applied voltage. When the sample was in the SmA phase the current was very small. For thick cells, for the applied voltage 2 V, the current started to increase and above 3.5 V the rate of increase in current dropped significantly. In thin cells at 45°C, increase in current dropped above 4 V. From microscopic observations, in the smectic phase, no flow of liquid was observed up to 10 V. So the electric current does not consist of any convective part in smectic phase. It has been shown experimentally [13] that in a liquid crystal under a dc field, the charge injection becomes significant in explaining instability such as FBI [5,6]. Combining it with electrical observations, one can easily conclude that after 2 V, charge injection took place and the diffusion current started to flow in addition to the ohmic part but no electric current due to fluid convection was present. As a result, above 3.5 V, the current tends to attain a constant value for thick cells. But in thin cells at 50°C and 53°C, the current continues to increase with applied voltage. Electric field in thin cells was higher than that for thick cells and it

may cause higher charge diffusion for the temperature close to the smectic-nematic phase transition temperature.

In thick cells, for nematic phase above 3 V and for isotropic liquid state above 2.5 V, the current was found to increase rapidly and in thin cells it was seen to grow just after 2 V. Similar behavior in I - V characteristics of 8CB and 8OCB in the nematic phase was observed by Rout and Choudhary [5,6] and in our previous work [7,8]. Above V_{th} , the force exerted on space charge was sufficient to start convection flow in the liquid crystal, the cellular domains appeared and the charge transfer with liquid flow was induced. As the voltage increases above V_{th} , the current due to charge transfer with convective flow of liquid becomes dominant and the total current increases very fast with voltage. When the liquid crystal was in isotropic phase, the same type of current-voltage characteristics was observed, however the current was higher.

(ii) Existence of charge diffusion :

To study the relaxation process, $I(t)$ - time (t) transient (where $I(t)$ is the current at the time, t , after switching off the supply voltage) traces of the sample were recorded in the

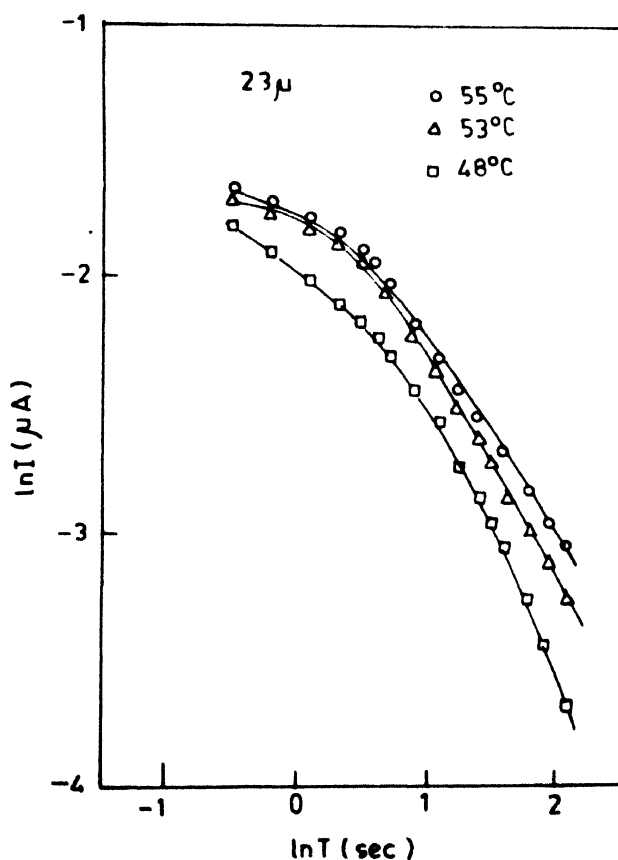


Figure 3(a). The plots of $\ln I(t)$ against $\ln t$ for 'S₃' for cell thickness $23 \mu m$.

memoryscope. This sample, S_3 , showed a slow relaxation. Previously Nakagawa [4], Rout and Choudhary [5,6] observed this type of slow relaxation in pure cyanobiphenyls. We found same type of relaxation in binary mixtures of cyanobiphenyls [7,8]. When a voltage

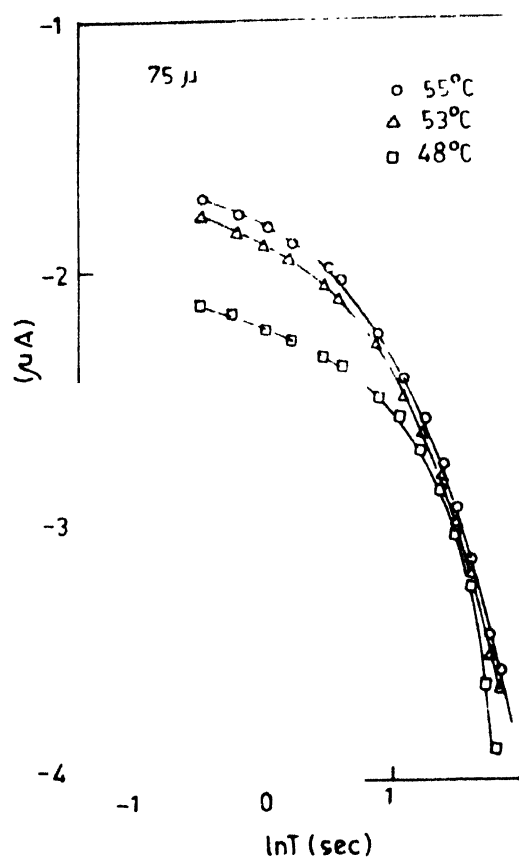


Figure 3(b). The plots of $\ln I(t)$ against $\ln t$ for ' S_3 ' for cell thickness $75 \mu\text{m}$.

of 2 V (less than V_{th} to avoid convection flow) was applied, the charges were injected into the liquid crystal from the electrodes, here from the negative electrode (Sb doped SnO_2 is a n-type semiconductor) and the charge started to diffuse into the liquid. After 4–5 minutes, the power supply was switched off and the diffused charges started to relax. An empirical formula $I(t) \propto (D/t)^{1/2}$ (where D is diffusion constant) for large values of t should be followed in the case of relaxation of diffused charges [5,6]. To examine the applicability and validity of this relation, $[\ln I(t) - \ln t]$ curves were drawn [Figures 3(a–b)]. It was observed that the relation holds good for $t > 2$ sec for the thick cells and $t > 1.6$ sec for thin cells. For $t < 2$ sec for the thick cells and $t < 1.6$ sec for thin cells, the current was found to deviate from the relation. It is due to director relaxation and also the approximations taken for large t was not suitable below the above limit. In the nematic phase, the diffusion was higher than that in the smectic phase. In the smectic phase, the diffusion of charges was temperature dependent. Rise in temperature causes a slight increase in diffusion of charge

in the nematic phase. In the temperature range between which smectic and nematic phase coexist showed the same type of increase in diffusion as seen in the nematic phase. No significant change was found in the case of the nematic to isotropic phase transition. For thin cell, above relation holds good for $t > 1.6$ sec which was due to the fact that higher electric fields help charges to diffuse into the liquid with higher amount. When the electric field was made zero, the contribution of the diffused charges to the current $I(t)$ was high, and within 1 sec, it started to dominate over director relaxation. In the smectic phase, charge diffusion was small and a large deviation from the diffusion relation was found. The slow molecular velocity in the smectic phase may retard the diffusion of charges in the smectic phase, particularly in the thick cells.

(iii) *Current-Temperature characteristics :*

Figures 4(a) and 4(b) show the current (I) – temperature (T) characteristics for different applied voltage. A sharp increase in electrical conductivity occurred within the temperature

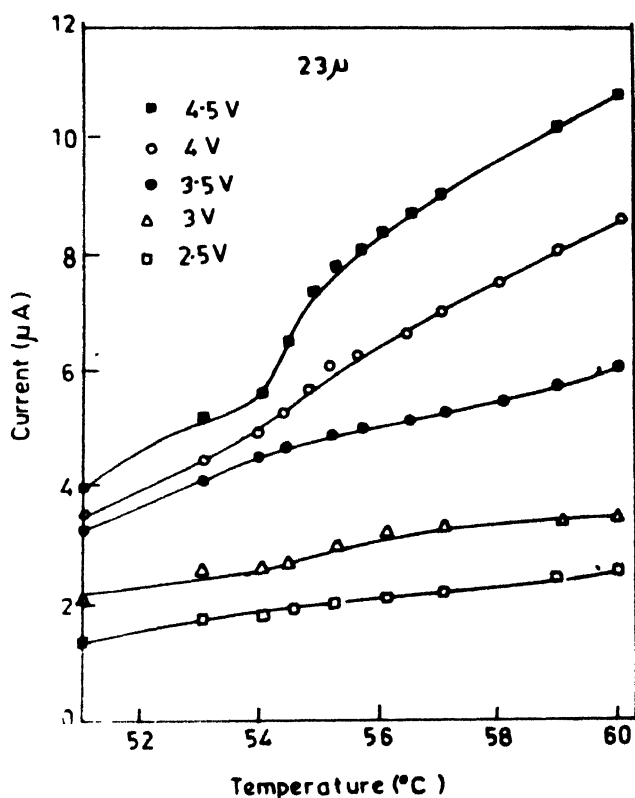


Figure 4(a). Current-temperature characteristics at different voltages for 'S₃' for cell thickness 23 μm.

range 53°C–54°C for thick cells [Figure 4(a) for 75 μm cell], but for thin cells [Figure 4(b) for 23 μm cell], the current in the liquid crystal in smectic phase was found to increase slowly with temperature at and above 3.5 V. In thin cells, in the temperature range 53.3°C–54°C (when the smectic and nematic phases coexist) the current did not increase with a rise

in the temperature. It started to increase rapidly within 54°C – 55°C . Above 55°C , the current grew with a smaller rate. In thick cells, the sharp rise in current was seen above 53.5°C , just

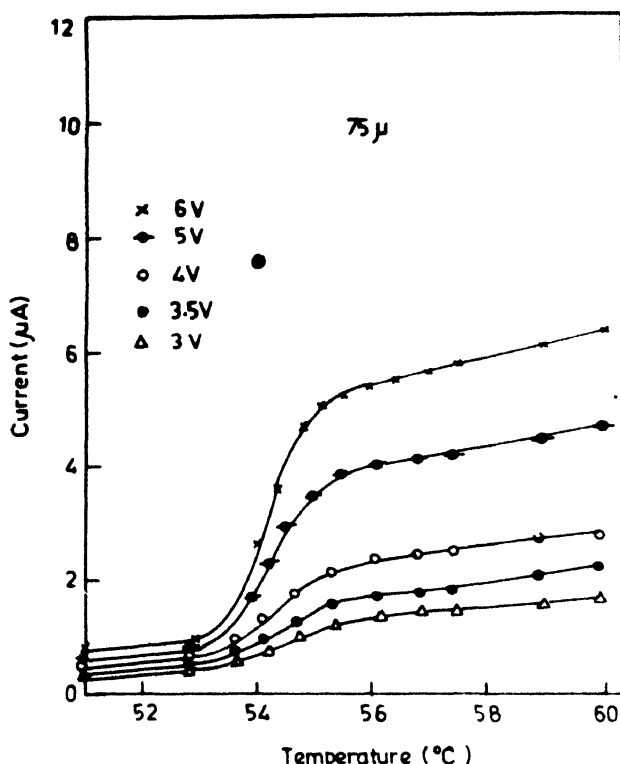


Figure 4(b). Current-temperature characteristics at different voltages for 'S₃' for cell thickness $75\ \mu\text{m}$.

after the starting of SmA-N phase transition and above 55°C the rate of increase of current became very small. In the temperature range 53.5°C – 55°C , SmA-N phase transition takes place, as a result the viscosity and elasticity change rapidly with temperature and an easier fluid flow occurs, the convective part of current becomes dominant. From the charge injection point of view Richardson emission's law was used and found useful to explain the current-temperature characteristics in the nematic phase for pure cyanobiphenyls [4–6]. However, in our previous [7,8] and present work, that equation was not so useful. Charge injection from the electrodes was the main reason for EHD instability but the flow of liquid during the instability determines the current. Viscosity [14], order parameter [11], elastic properties and molecular deformability [15] have been found strongly temperature dependent, and these play a major role in fluid flow. This was the main reason of the temperature dependence of electrical conductivity of the liquid crystal. It may be concluded that above 53.3°C phase transition in S₃ started and with increase in temperature, the nematic area within the cell increased, total amount of liquid taking part in convection increased resulting an increase in current with temperature. After completion of Sm-N phase transition (above 54°C) changes in viscosity, elastic property and other temperature

dependent physical parameters causes an increase in the flow rate of the fluid but above 55°C change in those physical parameters was not so much considerable.

It was also seen that at higher applied electric fields, the liquid suffered chemical dissociation and some empty areas developed within the cells specially for thin cells. Because of this problem, we could not take observations for higher electric fields. We were to restrict our electrical observations up to an applied voltage 5 V for thin cells and for thick cells the observations were conducted within 7 V.

4. Conclusions

- (i) Current flow in the liquid crystal mixture S_1 contains ohmic conduction, charge diffusion and charge transfer with fluid flow.
- (ii) There is no so much difference in EHD instability of the nematic and isotropic phase of the mixture S_3 .
- (iii) The liquid crystal mixture containing positive dielectric anisotropy and the splay elastic constant of the liquid crystal in the smectic A phase is higher than that in the nematic phase.

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